



TITLE:

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# Superconducting state coexisting with a phase-separated static magnetic order in (Ba,K)Fe<sub>2</sub>As<sub>2</sub>, (Sr,Na)Fe<sub>2</sub>As<sub>2</sub>, and CaFe<sub>2</sub>As<sub>2</sub>

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By muon spin-relaxation measurements on single-crystal specimens, we show that superconductivity in the AFe<sub>2</sub>As<sub>2</sub> (A=Ca,Ba,Sr) systems, in both the cases of composition and pressure tunings, coexists with a strong static magnetic order in a partial volume fraction. The superfluid response from the remaining paramagnetic volume fraction of (Ba<sub>0.5</sub>K<sub>0.5</sub>)Fe<sub>2</sub>As<sub>2</sub> exhibits a nearly linear variation in  $T$  at low temperatures, suggesting an anisotropic energy gap with line nodes and/or multigap effects.

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The announcement of superconductivity in La(O,F)FeAs ( $T_c=26$  K) (Ref. 1) triggered an unprecedented burst of research activities in FeAs-based superconductors and their parent systems. By now, superconductivity has been reported in systems with four different crystal structures, including the “1111” systems RE(O,F)FeAs with rare earth=La, Nd, Ce, etc.,<sup>2</sup> and the “122” systems AFe<sub>2</sub>As<sub>2</sub> (A=Ba, Sr, Ca).<sup>3–5</sup> Extensive measurements by neutron scattering,<sup>6–8</sup> Mössbauer effect,<sup>9–11</sup> and muon spin relaxation ( $\mu$ SR) (Refs. 11–15) have revealed collinear antiferromagnetic order in undoped parent compounds,<sup>6,16,17</sup> hyperfine splitting of <sup>57</sup>Fe Mössbauer spectra, and  $\mu$ SR frequencies indicative of a static moment size ranging between 0.3 and 0.8 Bohr magnetons per Fe,<sup>9,10,15</sup> and nearly linear scaling between  $T_c$  and the superfluid density<sup>12–14</sup> in the 1111 systems following the trend found in cuprate and other exotic superconductors.<sup>18</sup>

In studies of magnetic phase diagrams of the 1111 systems, as a function of increasing (O,F) substitution, La(O,F)FeAs shows<sup>19</sup> an abrupt and first-order-like evolution from an antiferromagnetic to superconducting state, Ce(O,F)FeAs shows nearly second-order-like evolution,<sup>7</sup> and Sm(O,F)FeAs (Ref. 20) exhibits phase-separated coexistence of static magnetism and superconductivity in a small concentration region around the phase boundary. Despite these differences, superconductivity appears mostly in the region without static magnetic order in the 1111 systems, similar to the case of the cuprates. In contrast, very little has been reported on the phase diagrams of the 122 systems. Recent powder neutron measurements on (Ba,K)Fe<sub>2</sub>As<sub>2</sub> (Ref. 8) found a phase diagram similar to the one for Sm(O,F)FeAs with co-

existing long-range magnetic order and superconductivity near the phase boundary, without providing information on the volume fraction of the magnetically ordered region. We have also reported  $\mu$ SR measurements on a single crystal of (Ba<sub>0.55</sub>K<sub>0.45</sub>)Fe<sub>2</sub>As<sub>2</sub> (Ref. 15) which found the coexistence of phase-separated static magnetic order and superconductivity. The superfluid density of this crystal was much lower than that in the corresponding 1111 systems with comparable  $T_c$ 's, which is suggestive of insufficient carrier doping. In the 122 systems, more definitive studies of magnetic phase diagrams can be expected due to the availability of large single crystals,<sup>21–23</sup> improvement of the growth method, and applicability of pressure tuning free from randomness due to substitution.

In this paper, we report  $\mu$ SR measurements of superconducting single crystals of (Ba<sub>0.5</sub>K<sub>0.5</sub>)Fe<sub>2</sub>As<sub>2</sub> ( $T_c \sim 37$  K) and (Sr<sub>0.5</sub>Na<sub>0.5</sub>)Fe<sub>2</sub>As<sub>2</sub> ( $T_c \sim 35$  K) in ambient pressure, and of CaFe<sub>2</sub>As<sub>2</sub> in ambient and applied pressure  $p$  up to  $p=10$  kbar, performed at TRIUMF in Vancouver, Canada. The former two crystals were prepared at the Institute of Physics in Beijing using the FeAs flux method<sup>23</sup> and weighed  $\sim 100$  and 40 mg, respectively. As shown in Fig. 1, sharp superconducting transitions were observed in the magnetic susceptibility of both crystals, which suggests that they are of good quality. No anomaly due to a structural transition can be seen in the resistivity results. These crystals were mounted with their  $ab$  planes perpendicular to the muon beam at the M20 channel. In our  $\mu$ SR measurements in transverse external fields applied perpendicular to the  $ab$  plane, both of these crystals exhibited a superfluid response

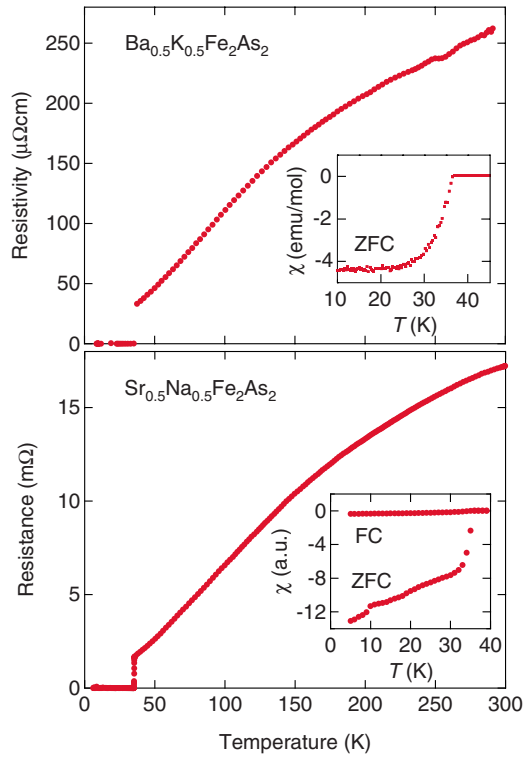


FIG. 1. (Color online) Temperature dependences of the resistivity and the magnetic susceptibility of our specimens of (a)  $(\text{Ba}_{0.5}\text{K}_{0.5})\text{Fe}_2\text{As}_2$  ( $T_c \sim 37$  K) and (b)  $(\text{Sr}_{0.5}\text{Na}_{0.5})\text{Fe}_2\text{As}_2$  ( $T_c \sim 35$  K). Due to the irregular shape of the specimen which prevents accurate estimate of the demagnetizing factor, we put the results of the latter system on a relative (arbitrary) scale. The inset figure of (b) shows the magnetic susceptibility obtained in the field-cooling and zero-field-cooling procedures.

corresponding to a muon spin-relaxation rate of  $\sim 1 \mu\text{s}^{-1}$  at  $T \rightarrow 0$ . This indicates strong bulk superconductivity of these crystals.

Over 1 g of  $\text{CaFe}_2\text{As}_2$  crystals (in more than 100 pieces), prepared in Ames Laboratory using the Sn flux method,<sup>22</sup> were mounted in a pressure cell having a sample space of 7 mm in diameter and 12 mm long. The cell was pressurized at room temperature before being mounted in the cryostat. Daphne oil was used as the pressure mediator. This was chosen because it is known not to solidify at room temperature up to  $\sim 20$  kbar and so generates hydrostatic pressure over a wider pressure temperature range than Fluorinert, which solidifies above  $\sim 10$  kbar at room temperature. The crystals were aligned with their *ab* planes perpendicular to the muon beam at the M9B channel, where the initial muon spin polarization is tuned to be perpendicular to the beam direction.

$\mu\text{SR}$  measurements were performed in zero field (ZF) and weak transverse field (WTF) of  $\sim 30$ – $50$  G to study magnetic ordering, and in transverse field (TF) of  $300$ – $500$  G to study superfluid density. A recent study on  $(\text{Ca}, \text{Sr})\text{RuO}_3$  and  $\text{MnSi}$  in applied pressure<sup>24</sup> has demonstrated  $\mu\text{SR}$ 's unique capability of determining volume fractions of regions with and without static magnetic order in systems having phase separation. Details of the  $\mu\text{SR}$  methods can be found in Refs. 24 and 25.

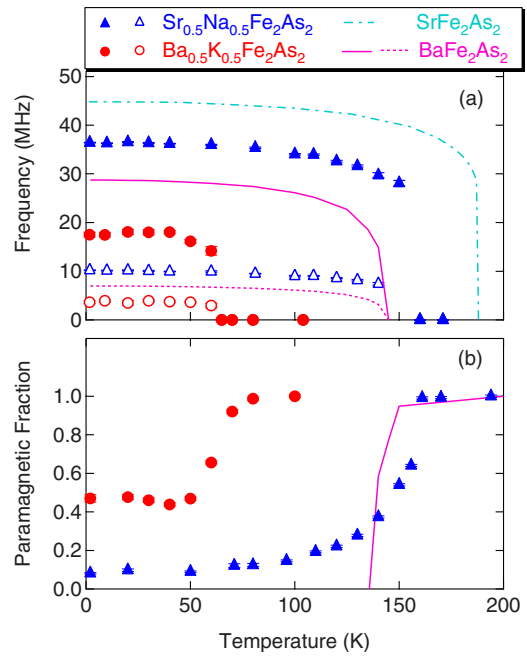


FIG. 2. (Color online) Temperature dependences of (a) the muon spin precession frequency observed in ZF  $\mu\text{SR}$  and (b) the paramagnetic volume fraction determined from WTF- $\mu\text{SR}$  measurements of single-crystal specimens of  $(\text{Ba}_{0.5}\text{K}_{0.5})\text{Fe}_2\text{As}_2$  ( $T_c \sim 37$  K) and  $(\text{Sr}_{0.5}\text{Na}_{0.5})\text{Fe}_2\text{As}_2$  ( $T_c \sim 35$  K). The results from the present work (solid and open symbols) are compared with those of the undoped parent compounds  $\text{BaFe}_2\text{As}_2$  (Ref. 15) and  $\text{SrFe}_2\text{As}_2$  (Ref. 10) (solid and broken lines).

Figure 2 shows (a) the muon spin precession frequencies observed in ZF  $\mu\text{SR}$  and (b) the paramagnetic volume fraction derived from WTF- $\mu\text{SR}$  measurements in the  $(\text{Ba}, \text{K})$  and  $(\text{Sr}, \text{Na})$  crystals. The solid lines show the reported results in the undoped parent compounds  $\text{BaFe}_2\text{As}_2$  (Ref. 15) and  $\text{SrFe}_2\text{As}_2$ .<sup>10</sup> In both systems, static magnetism sets in at temperatures well above the superconducting  $T_c$ 's, in a large volume fraction of  $\sim 90\%$  in the  $(\text{Sr}, \text{Na})$  system and  $50\%$  in the  $(\text{Ba}, \text{K})$  system. We observed two different precession frequencies in a given system, presumably coming from two different muon sites, as was the case in  $\text{BaFe}_2\text{As}_2$ .<sup>15</sup> The frequencies in the superconducting samples are reduced from the values in the undoped compounds only by  $20$ – $30\%$ , indicating that static magnetic order with a significant Fe moment size exists in the magnetically ordered regions. These results demonstrate phase separation between magnetically ordered and paramagnetic volumes.

In TF  $\mu\text{SR}$ , the precession signal from the paramagnetic volume fraction exhibits damping below  $T_c$  due to an inhomogeneous field distribution in the flux vortex lattice. The relaxation rate  $\sigma$ , obtained by fitting the spectra to a Gaussian function  $\exp(-\sigma^2 t^2/2)$ , is given by  $\sigma \propto 1/\lambda^2 \propto n_s/m^*$ , where  $\lambda$  is the penetration depth,  $n_s$  is the superconducting carrier density, and  $m^*$  is the effective mass.<sup>18,25</sup> An increase in  $\sigma$  was observed in both the  $(\text{Ba}, \text{K})$  and  $(\text{Sr}, \text{Na})$  crystals below the superconducting  $T_c$ 's. Since the statistical accuracy of the data is much better for the former system with the larger paramagnetic volume fraction, here we present the results of  $(\text{Ba}_{0.5}\text{K}_{0.5})\text{Fe}_2\text{As}_2$  in TF =  $500$  G applied parallel to

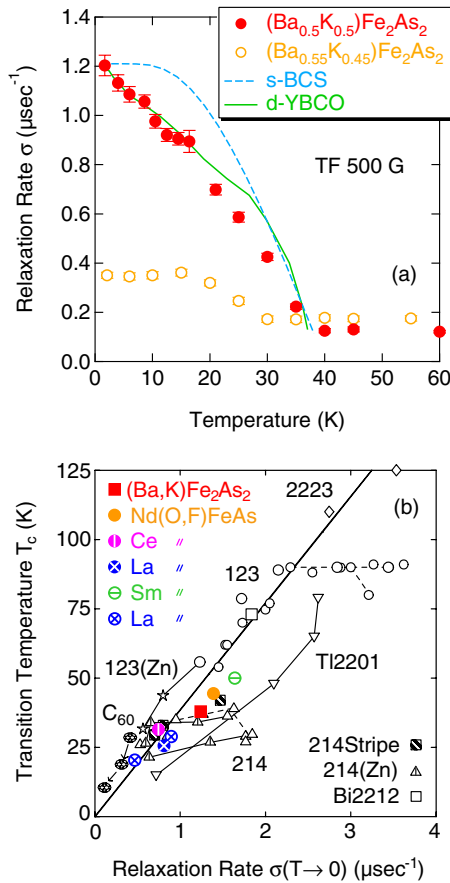


FIG. 3. (Color online) (a) Temperature dependence of the muon spin-relaxation rate  $\sigma$  observed in a single-crystal specimen of  $(\text{Ba}_{0.5}\text{K}_{0.5})\text{Fe}_2\text{As}_2$  ( $T_c \sim 37$  K) in TF  $\mu\text{SR}$  with TF=500 G (closed circles, present work), compared with the temperature dependence expected for the isotropic energy gap of BCS  $s$ -wave pairing (broken line), scaled results from YBCO (Ref. 25) (solid line), and our previous results in a different crystal of  $\text{Ba}_{0.55}\text{K}_{0.45}\text{Fe}_2\text{As}_2$  (Ref. 15) (open circles). (b) A plot of the relaxation rate  $\sigma(T \rightarrow 0)$  versus  $T_c$ , including the point for  $(\text{Ba}_{0.5}\text{K}_{0.5})\text{Fe}_2\text{As}_2$  from the present work as well as those for the FeAs-based 1111 superconductors published in Refs. 12 and 14, various cuprates, and  $\text{A}_3\text{C}_{60}$  superconductors (Ref. 18).

the  $c$  axis in Figs. 3(a) and 3(b). The temperature dependence of  $\sigma$  in (a) is nearly linear with  $T$ , as demonstrated by the good agreement with the scaled data from a  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  (YBCO) system.<sup>25</sup> The observed behavior is distinctly different from the case for an isotropic energy gap shown by the broken line representing a calculation for BCS  $s$ -wave coupling. The observed temperature dependence may be attributed to (1) line nodes in an anisotropic energy gap or (2) widely different magnitudes of multiple isotropic gaps as seen in calculations<sup>26,27</sup> based on multiple bands. An angle resolved photoemission spectroscopy (ARPES) measurement<sup>28</sup> on  $(\text{Ba},\text{K})\text{Fe}_2\text{As}_2$  reported evidence for isotropic multiple gaps, consistent with theories based on an extended  $s$ -wave pairing.<sup>29,30</sup>

The absolute value of  $\sigma(T \rightarrow 0)$  is about a factor of 3 larger than that observed in  $(\text{Ba}_{0.55}\text{K}_{0.45})\text{Fe}_2\text{As}_2$  in our previous measurements.<sup>15</sup> Given that the  $H_{c2}$  anisotropy is rela-

tively low near  $T_c$ , varying between 3.5 and 2.5 for  $H < 14$  T (Ref. 21) and decreasing for higher fields,<sup>31</sup> we plot the present results in the  $\sigma(T \rightarrow 0)$  versus  $T_c$  plot in Fig. 3(b) without single crystal to polycrystalline conversion corrections.<sup>32</sup> The resulting point from the present  $(\text{Ba},\text{K})$  crystal (red solid square symbol) indicates that the present system has a superfluid density close to those in the 1111 systems with comparable  $T_c$ 's and that a sufficiently doped 122 FeAs system follows the nearly linear relationship between  $T_c$  and  $n_s/m^*$  found in the cuprates and 1111 systems.

The superconducting state can also be obtained by applying pressure (using a liquid pressure medium) to the undoped parent compounds of the 122 systems.<sup>4,5</sup> In particular, the  $\text{CaFe}_2\text{As}_2$  system shows superconductivity below  $T \sim 10$  K at relatively low pressures  $p$  of 3–8 kbar, which are attainable using the available  $\mu\text{SR}$  piston-cylinder pressure cell. We studied static magnetic order of  $\text{CaFe}_2\text{As}_2$  at ambient pressure and at  $p=3.9$ , 6.2, and 9.9 kbar by performing WTF  $\mu\text{SR}$  with WTF=50 G. Solid symbols in Fig. 4(a) show the paramagnetic volume fraction, obtained after subtracting the contribution from the pressure cell in which the single-crystal specimens were placed with their  $c$  axis oriented parallel to the beam direction. Open circle symbols represent additional results in ambient pressure obtained without the pressure cell. Figure 4(a) demonstrates that static magnetic order sets in at temperatures well above the superconducting  $T_c$  in a partial volume fraction both at  $p=3.9$  and 6.2 kbar. The static magnetism disappears at  $p=9.9$  kbar, where the superconducting state no longer exists. Figure 4(b) shows the low-temperature ( $T \rightarrow 0$ ) values of the volume fraction of the magnetically ordered region (from WTF data) as well as the muon spin precession frequency in ZF  $\mu\text{SR}$ , which is proportional to the size of the ordered Fe moment. We present the resulting pressure-temperature phase diagram of  $\text{CaFe}_2\text{As}_2$  in Fig. 4(c). The superconducting phase boundary in this figure is based on the reported resistivity results.<sup>4</sup> Figure 4 indicates that a rather strong magnetism exists in a substantial volume fraction below  $T=50$ –100 K, which is well above the superconducting  $T_c$ , as in the cases of the  $(\text{Ba},\text{K})$  and  $(\text{Sr},\text{Na})$  crystals at ambient pressure.

Resistivity<sup>4</sup> and neutron<sup>33</sup> measurements in applied pressure, the former (the latter) using Fluorinert (He gas) as the pressure mediator, have been reported on  $\text{CaFe}_2\text{As}_2$  single crystals prepared by an identical method to that used for the present specimens.<sup>22</sup> In the resistivity studies, a sharp jump was observed at  $T=170$  K at ambient pressure, corresponding to the first-order tetragonal-to-orthorhombic structural phase transition below which magnetic order was detected both by neutrons and muons. With increasing pressure this feature broadens and ordering moves toward lower temperatures, which is qualitatively consistent with the present results in Fig. 4(c). The resistivity anomaly disappears above  $p \sim 4$  kbar, and the neutron magnetic Bragg-peak intensity at  $T=50$  K becomes nearly equal to the background level at  $p=6.3$  kbar [Fig. 1(c) in Ref. 33], while  $\mu\text{SR}$  detected magnetic order continuing to exist at  $p=6.2$  kbar, albeit in a partial volume fraction.

Quite recently, additional neutron measurements under pressure, using Fluorinert (and not He gas) as the pressure mediator, were performed<sup>34</sup> to examine  $\text{CaFe}_2\text{As}_2$  specimens



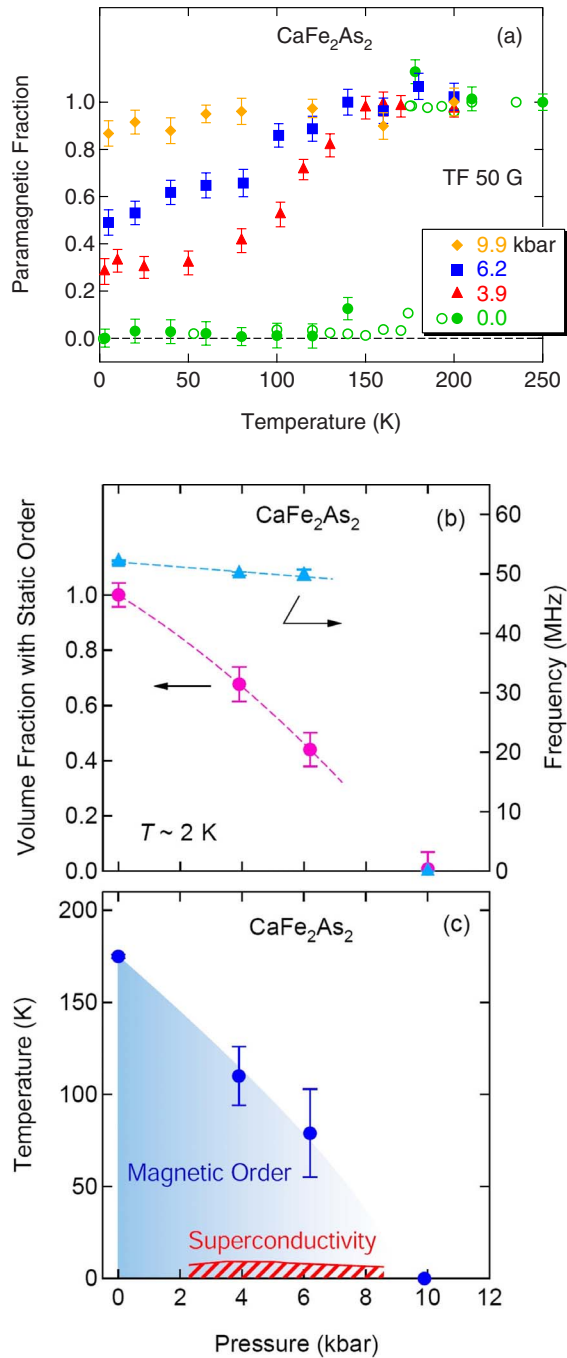


FIG. 4. (Color online) (a) The volume fraction of regions without static magnetic order in  $\text{CaFe}_2\text{As}_2$ , as a function of temperature and pressure, determined by WTF- $\mu$ SR measurements with WTF  $\sim 50$  G. The points with closed (open) symbols were obtained in measurements with (without) a pressure cell. (b) Pressure dependence of the volume fraction of the magnetically ordered region (purple closed circles; left axis) from WTF  $\mu$ SR and the muon spin precession frequency (blue triangles; right axis) from ZF  $\mu$ SR at  $T \sim 2$  K in  $\text{CaFe}_2\text{As}_2$ . (c) The phase diagram as a function of pressure and temperature in  $\text{CaFe}_2\text{As}_2$ . The  $T_c$  values are taken from the reported resistivity results (Ref. 4). Upper, middle, and lower temperatures attached to the closed circle symbols for  $T_N$  represent temperatures at which the volume fraction with static magnetic order becomes 30%, 50%, and 70% of the value at  $T \rightarrow 0$ , respectively.

grown by the same group<sup>22</sup> as those discussed in the present work. These neutron measurements revealed the coexistence of two structural phases (antiferromagnetic orthorhombic and nonmagnetic collapsed tetragonal phases) at low temperatures, which is also consistent with our  $\mu$ SR results. This behavior probably arises from the applied pressure not actually being truly hydrostatic, as  $\text{CaFe}_2\text{As}_2$  seems to be strongly affected by any slight deviation from hydrostaticity. More specifically, superconductivity was not observed in resistivity and susceptibility measurements<sup>35</sup> of  $\text{CaFe}_2\text{As}_2$  under He gas pressure, which is capable of providing true hydrostatic pressure over a much wider pressure temperature range than either Fluorinert or Daphne oil. The crystals investigated by Yu *et al.* were also grown by the same group<sup>22</sup> as those studied in the present work. Superconductivity, the coexistence of magnetically ordered and nonmagnetic fractions and the coexistence of the orthorhombic and collapsed tetragonal structures appear only under pressure using a liquid pressure medium. The magnetic order in a partial volume fraction may play an important role in the emergence of superconductivity in  $\text{CaFe}_2\text{As}_2$ .

The present  $\mu$ SR results do not provide direct evidence for distinguishing whether superconductivity lives uniformly over the entire volume or exclusively in the paramagnetic volume fraction. Evidence for the latter case has been reported in ARPES studies on a lightly doped single crystal of  $(\text{Ba},\text{K})\text{Fe}_2\text{As}_2$ ,<sup>36</sup> which found an intensity ratio of an ungapped magnetic response to a gapped superconducting response comparable to the ratio of volumes with and without static magnetic order found by  $\mu$ SR. This situation is similar to the case of  $(\text{La},\text{Eu},\text{Sr})_2\text{CuO}_4$  where the static volume fraction and superfluid density in  $\mu$ SR exhibit a tradeoff.<sup>37</sup> The present study does not provide an estimate of the length scale of the phase-separated regions. In  $\text{La}_2\text{CuO}_{4.11}$ ,<sup>38</sup> we estimated this length to be a few nanometers.

Phase separation at the border of magnetic and superconducting states has been found also in the organic  $(\text{BEDT-TTF})_2\text{-X}$  superconductors<sup>39</sup> and  $\text{CeCu}_2\text{Si}_2$ .<sup>40</sup> First-order phase transitions, similar to those in the La1111 FeAs system,<sup>19</sup> have been found in the  $\text{A}_3\text{C}_{60}$ ,<sup>41</sup>  $\text{Ce}(\text{Co},\text{Cd})\text{In}_5$ , and  $\text{CeRhIn}_5$  (Ref. 42) systems. Phase separation was also discovered at the border of a collinear antiferromagnetic state and a nonmagnetic spin-gap state in an insulating  $J_1$ - $J_2$  spin system  $\text{Cu}(\text{Cl},\text{Br})\text{La}(\text{Nb},\text{Ta})_2\text{O}_7$ ,<sup>43</sup> and at the border of an itinerant heli/ferromagnetic state and paramagnetic state in  $\text{MnSi}$  and  $(\text{Sr},\text{Ca})\text{RuO}_3$ .<sup>24</sup> Further exploration of these behaviors will lead to a better understanding of superconductivity and magnetism in correlated-electron systems.

Regarding the pairing symmetry, available experimental results on the 1111 and 122 systems are divided between those favoring an isotropic nodeless gap<sup>44</sup> and those supporting line nodes and/or multigap features.<sup>26,27,45</sup> The  $(\text{Ba}_{0.5}\text{K}_{0.5})\text{Fe}_2\text{As}_2$  result in Fig. 3(a) has established at least one definite case which does not agree with a single isotropic energy gap. The different temperature dependences of the superfluid density between the two  $(\text{Ba},\text{K})\text{Fe}_2\text{As}_2$  crystals in Fig. 3(a) may be due to a doping dependence of multiple gap magnitudes, as proposed in Ref. 30. The nearly linear relationship between  $T_c$  and the superfluid density [Fig. 3(b)] followed by cuprates, 1111 FeAs, 122 FeAs, and  $\text{A}_3\text{C}_{60}$  sys-

tems suggests the existence of an underlying generic principle common to the condensation mechanisms of all these exotic superconductors.<sup>18,46</sup>

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